Finite temperature Mott transition in Hubbard model on anisotropic triangular lattice

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We investigate the Hubbard model on the anisotropic triangular lattice by means of the cellular dynamical mean field theory. The phase diagram determined in the Hubbard interaction versus temperature plane shows novel reentrant behavior in the Mott transition due to the competition between Fermi-liquid formation and magnetic correlations under geometrical frustration. We demonstrate that the reentrant behavior is characteristic of the Mott transition with intermediate geometrical frustration and indeed consistent with recent experimental results of organic materials.

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Geometrical frustration has attracted much interest in the field of strongly correlated electron systems. The discovery of heavy fermion behavior in the pyrochlore compound LiV_2O_4 [1] and the superconductivity in the triangular lattice compound $Na_xCoO_2 \cdot yH_2O$ [2] have stimulated intensive studies of frustrated electron systems. Geometrical frustration has also uncovered new aspects of the Mott metal-insulator transition, which is now one of the central issues in the physics of strongly correlated electron systems. In particular, recent experiments on the triangular lattice organic materials κ-(BEDT-TTF)$_2$X have revealed various interesting physics, such as a novel spin liquid state in the Mott insulating phase, unconventional superconductivity, etc [3, 4, 5]. Since the path integral renormalization group study of the triangular lattice Hubbard model [6], the correlated electrons on the anisotropic triangular lattice have been intensively studied so far [7, 8, 9, 10, 11, 12, 13, 14, 15, 16]. Geometrical frustration effects on the finite-temperature (T) Mott transition, however, have not been sufficiently understood yet. One of interesting and nontrivial features in the finite-T Mott transition is reentrant behavior observed in the frustrated organic material κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl under pressure [3, 5]. With lowering temperature, it once undergoes a transition from Mott insulator to metal, and then reenters the *paramagnetic* insulating phase at a much lower temperature. This reentrant behavior is quite different from the Mott transition in the three dimensional systems, such as V_2O_3, and is expected to be a new aspect due to the geometrical frustration and/or low-dimensionality.

The dynamical mean field theory (DMFT) has given substantial theoretical progress in understanding the Mott transition, but this method, not treating spatial fluctuations, does not explain the reentrant behavior: In DMFT, the metallic phase always lies on the lower temperature side of the finite-T Mott transition line [18]. It is thus desirable to properly incorporate spatially extended correlations and geometrical frustration by employing a different appropriate method.

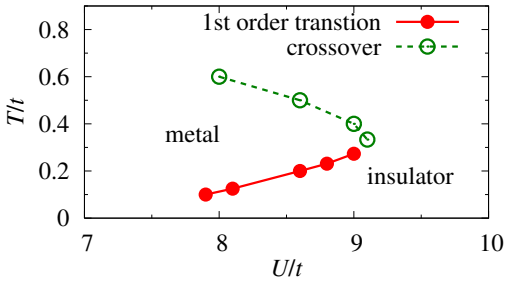

FIG. 1: Phase diagram of Hubbard model on anisotropic triangular lattice for $t'/t = 0.8$.

In this paper, we study the finite-T Mott transition in the Hubbard model on the anisotropic triangular lattice by means of a cluster extension of DMFT, the cellular-DMFT (CDMFT) [19, 20]. The obtained phase diagram indeed shows a reentrant behavior in the Mott transition (see Fig. 1), which is consistent with that observed in the organic material mentioned above. We argue that the reentrant behavior is a generic feature inherent in the Mott transition with intermediate geometrical frustration, and thus can be found in various frustrated electron systems experimentally.

We consider the standard Hubbard model on the triangular lattice with the hopping anisotropy (see Fig. 2(a)),

$$H = -t \sum_{\langle i,j\rangle,\sigma} c^\dagger_{i\sigma} c_{j\sigma} - t' \sum_{(i,j),\sigma} c^\dagger_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}, \quad (1)$$

with $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$, where $c^\dagger_{i\sigma}$ ($c_{j\sigma}$) creates (annihilates) an electron with spin σ at site i. To analyze this model

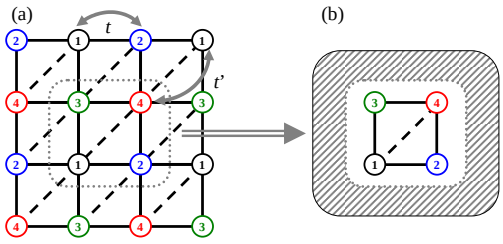

FIG. 2: (a) Sketch of the anisotropic triangular lattice and (b) the effective cluster model using four sites cluster CDMFT.

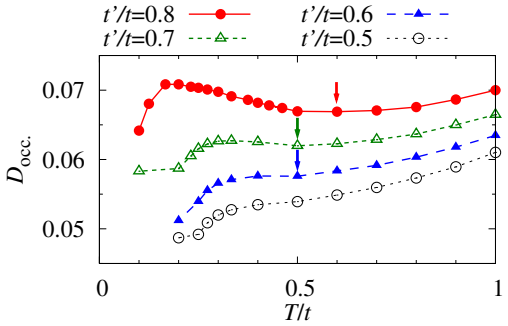

FIG. 3: Nonmonotonic temperature dependence of double occupancy for $U/t = 8.0$, where ground states are insulating and in the vicinity of the Mott transition. Arrows denote the crossover temperature from the high-T insulator to the intermediate-T metal.

we use CDMFT, which has been successfully applied to frustrated systems such as the Hubbard model on the triangular lattice [11, 15, 17] and the Kagomé lattice [21]. In CDMFT, the original lattice is regarded as a superlattice consisting of clusters, which is then mapped onto an effective cluster model via a standard DMFT procedure. Considering four sublattices labeled by 1-4, as shown in Fig. 2(a), the original lattice model is converted into a four-site cluster model coupled to the self-consistently determined medium illustrated in Fig. 2(b). Given the Green's function for the effective medium, $\hat{\mathcal{G}}$, we can compute the cluster Green's function $\hat{G}$ and the cluster self-energy $\hat{\Sigma}$ by simulating the effective cluster model with Quantum Monte Carlo (QMC) method [23]. Here, $\hat{\mathcal{G}}$, $\hat{G}$, and $\hat{\Sigma}$ are described by 4×4 matrices. The effective medium $\hat{\mathcal{G}}$ is then computed via the Dyson equation,

$$\hat{\mathcal{G}}^{-1}(\omega) = \left[\sum_{\mathbf{K}} \frac{1}{\omega + \mu - \hat{t}(\mathbf{K}) - \hat{\Sigma}(\omega)}\right]^{-1} + \hat{\Sigma}(\omega), \quad (2)$$

where μ is the chemical potential and $\hat{t}(\mathbf{K})$ is the Fourier-transformed hopping matrix for the sublattice. Here, summation of $\mathbf{K}$ is taken over the reduced Brillouin zone. We repeat this procedure until the results are converged (fifty times iterations are enough). In each iteration, we typically use 10^6 QMC sweeps and Trotter time slices $L = 12t/T$, where time slice errors are reduced by interpolation scheme based on a high-frequency expansion of Green's function [24].

Let us now study the finite-T Mott transition in the model (1) at half filling. We first investigate the T-dependence of the double occupancy $D_{\mathrm{occ.}} = \langle n_{i\uparrow} n_{i\downarrow} \rangle$ for interaction strength $U/t = 8$, where the ground state is insulating and in the vicinity of Mott transition point. A remarkable property in our frustrated system is nonmonotonic T-dependence of $D_{\mathrm{occ.}}$. As shown in Fig. 3, $D_{\mathrm{occ.}}$ decreases at high-T, and then shows an upturn at intermediate-T taking a local minimum at $T/t \sim 0.5$, as T decreases. At much lower T, $D_{\mathrm{occ.}}$ starts to decrease again, showing a hump structure. This nonmonotonic T-dependence of $D_{\mathrm{occ.}}$ indicates that the system once changes from insulating to metallic and then reenters the insulating phase as T is lowered. The behavior is quite different from that in the infinite dimensional Hubbard model, in which $D_{\mathrm{occ.}}(T)$ has a single minimum at the Fermi-liquid (FL) coherence temperature T_0. In the latter case, the system is insulating at $T > T_0$ and FL at $T < T_0$ [18]. The nonmonotonic behavior in our system is also different from that in the unfrustrated square lattice Hubbard model. On the square lattice, the FL coherence is disturbed by the antiferromagnetic (AF) interaction due to the perfect nesting, which gives rise to monotonic decrease of $D_{\mathrm{occ.}}$ [25, 26]. Note that the hump structures in $D_{occ.}$ become more prominent and shift to lower temperatures as t' increases, although they get smeared for $t'/t = 0.5$. Therefore, the nonmonotonic behavior is a characteristic feature caused by geometrical frustration.

To elucidate whether the change between metal and insulator is a real phase transition or crossover, we further investigate the double occupancy for typical anisotropy $t'/t = 0.8$ with varying U. We start from the non-interacting system to reach the large-U regime, typically $U/t = 10$, and then calculate $D_{\mathrm{occ.}}$ with decreasing U gradually. As shown in Fig. 4, the double occupancy jumps at critical interaction strength U_c with decrease of U, which signals the first order Mott transition. The jump shrinks with increasing T, and vanishes above $T/t \sim 0.3$. The critical end point is expected to be located at $T/t \sim 0.3$ and $U/t \sim 9.1$. At high-T, the system shows a crossover between metal and insulator, where we define the boundary by the temperature at which the double occupancy takes the first local minimum at high-T (see Fig. 3). This boundary is consistent with that determined by the local minimum of the density of states at the Fermi energy. The phase diagram thus determined is drawn in Fig. 1. The remarkable point is that $U_c(T)$ in our system shows a slope with the opposite sign to that in the infinite dimensional Hubbard model at low temperatures, whereas the high-T crossover shows similar behavior to the well-known results in infinite dimensions.

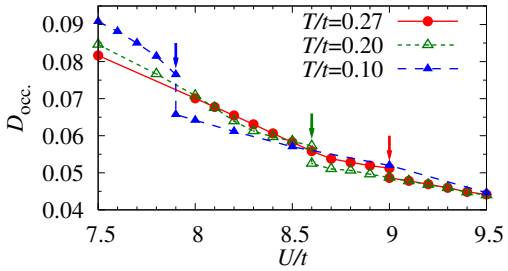

FIG. 4: Double occupancy as a function of interaction strength U/t for $t'/t = 0.8$ at several temperatures. We show only the transition from insulator to metal with weakening U (defined as U_{c1}), although we find hysteresis.

Within the single-site DMFT, entropy per site in the paramagnetic insulating phase is roughly $\ln 2$ corresponding to localized free spins and the metallic phase with the FL coherence has smaller entropy. Hence, in the vicinity of Mott transition temperature, the system becomes insulating at high-T to gain the entropy and the metallic phase always lies in the lower temperature regime [18]. However, the spatial fluctuations, which are not incorporated in the single-site DMFT, become important at low temperatures. For example, the dynamical cluster treatment of the unfrustrated square lattice shows that the FL metallic phase does not appear because of strong AF correlations [25, 26]. On the other hand, in our system, the magnetic correlations are hard to develop down to low temperature $T/t \sim 0.4$ because of geometrical frustration. Therefore, as temperature decreases, the entropy is released not by spin correlations but by the itinerancy of electrons in $T/t > 0.4$, which causes the crossover from insulator to metal in Fig. 1. The appearance of this kind of FL states is one of characteristics near the Mott transition with geometrical frustration [21]. At much lower temperatures, the magnetic correlations get enhanced, which finally trigger the first order transition from the FL to the insulator with smaller entropy. Therefore, as T decreases, $U_c(T)$ decreases at low temperatures ($T/t < 0.4$) in contrast to at high temperatures ($T/t < 0.4$). We thus conclude that the reentrant Mott transition on the anisotropic triangular lattice is due to the competition induced by geometrical frustration between the FL formation and the magnetic correlations. Since such competition is expected to happen in frustrated electron systems in low dimensions, the reentrant behavior in the Mott transition could be found in a wide variety of frustrated materials.

We can clearly see the development of the quasiparticles and magnetic correlations discussed above in the momentum resolved one-particle spectrum $A_{\mathbf{k}}(\omega)$. Within CDMFT, the one-particle Green's function is given as,

$$G_{\mathbf{k}}(\omega) = \frac{1}{4}\sum_{\gamma,\delta} e^{i\mathbf{k}\cdot(\mathbf{r}_\gamma - \mathbf{r}_\delta)} \left[\omega + \mu - \hat{t}(\mathbf{k}) - \hat{\Sigma}(\omega)\right]^{-1}_{\gamma\delta}, \quad (3)$$

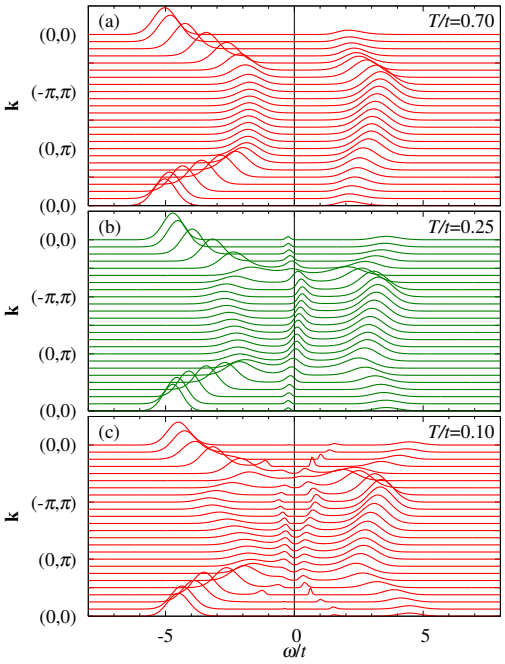

FIG. 5: Momentum resolved one-particle spectrum $A_{\mathbf{k}}(\omega)$ for $U/t = 8.0$, $t'/t = 0.8$ at several temperatures.

where $\mathbf{k}$ is the wave vector in the original Brillouin zone and $\mathbf{r}_\gamma$, $\mathbf{r}_\delta$ label four cluster sites [22]. We calculate the imaginary time Green's function $G_{\mathbf{k}}(\tau)$ and obtain the spectrum $A_{\mathbf{k}}(\omega) = -\mathrm{Im}G_{\mathbf{k}}(\omega + i0)/\pi$ using the maximum entropy method [27]. In Fig. 5, we show $A_{\mathbf{k}}(\omega)$ for $U/t = 8$, $t'/t = 0.8$ at typical temperatures. At high temperatures $A_{\mathbf{k}}(\omega)$ shows an insulating behavior, where it has a large Hubbard gap of order of U/t and no quasiparticle peak. As temperature is lowered, the quasiparticle peak starts to develop inside the gap, having weak dispersion, which clearly demonstrates the emergence of the frustration-induced metallic phase. The stabilization of the metallic phase by geometrical frustration is consistent with the previous studies of the Hubbard model on the triangular lattice [8, 26] and the Kagomé lattice [21]. At much lower temperatures, the quasiparticle peaks split and acquire a very small gap, where the system is insulating again. This small gap is due to the magnetic exchange interaction among the quasiparticles, which is consistent with the results at zero temperature recently obtained by CDMFT with exact diagonalization method [15]. We thus confirm that our frustrated system exhibits the insulator-metal-insulator reentrant behavior as temperature decreases.

Finally we examine the magnetic instability by calculating the static spin susceptibility χ_q. We find that the spin susceptibility χ_q remains finite just below the first order Mott transition temperature and hence the *paramagnetic* Mott insulator is not precluded by the magnetically ordered phase in contrast to the single-site DMFT

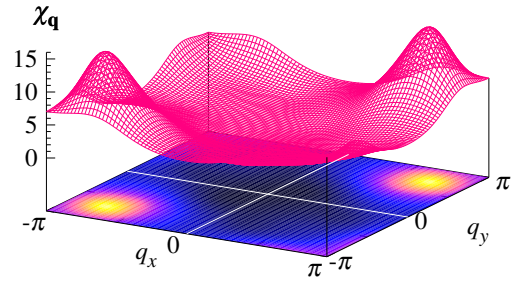

FIG. 6: Static spin susceptibility χ_q in the insulating phase for $U/t = 9$, $t'/t = 0.8$ at $T/t = 0.2$.

where the magnetic order almost conceals the Mott transition [18, 28]. Although the appearance of a finite-temperature magnetic transition is due to a mean-field type approximation, the magnetic ordering in our case, highly suppressed by frustration effects, appears below the Mott transition temperature. In Fig. 6, we show χ_q for $U/t = 9$, $t'/t = 0.8$ at $T/t = 0.2$, where the system is in the insulating phase close to the Mott transition point. The susceptibility χ_q takes a maximum at incommensurate wave vectors $\mathbf{q} \sim (0.7\pi, 0.7\pi)$ and does not diverge. The magnetic correlations thus play an important role in driving the Mott transition at low temperatures, but do not induce a real magnetic instability due to strong frustration. We note that our calculations of χ_q for different values of t' are also consistent with recent results of the Hubbard model around the Mott transition [6, 7, 13, 14]: For large-U and weak frustration $t'/t \leq 0.7$, χ_q has a peak at $\mathbf{q} = (\pi, \pi)$ corresponding to the commensurate antiferromagnetism, which rapidly develops with lowering T. We also find that χ_q for $t'/t = 1$ takes a maximum at $\mathbf{q} = (2\pi/3, 2\pi/3)$ corresponding to the 120° structure, in which the development of χ_q is much slower than the AF correlations for small t'.

In summary, we have investigated the Mott transition in the Hubbard model on the anisotropic triangular lattice by means of CDMFT. The obtained phase diagram (Fig. 1) in the U-T plane for $t'/t = 0.8$ is qualitatively consistent with the reentrant Mott transition in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl with $t'/t \sim 0.75$. By calculating the wave-vector dependent spectral function and susceptibility, we have clarified that the reentrant-type Mott transition is due to the competition between the FL formation and the magnetic correlations under geometrical frustration. We have also studied the Mott transition for different values of t'/t. For weakly frustrated case $t'/t \leq 0.5$, we have found that the FL formation is suppressed by the strong AF correlations, so that the reentrant Mott transition becomes obscured. On the other hand, we have found that for fully frustrated case $t' \sim t$, the FL states are well stabilized by frustration and the metallic region is extended, because the magnetic fluctuations of the 120° structure are weak. Hence, the low-T Mott transition line shifts to much lower-T regime [11]. The above tendency is consistent with another organic material κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ with $t' \sim t$, where magnetic ordering was not observed down to the lowest temperature studied experimentally. Therefore the reentrant behavior can be most clearly observed in moderately frustrated electron systems. Although we have used the four-site-cluster CDMFT, we believe that the reentrant behavior of Mott transition is robust and our results do not qualitatively change even for a larger cluster size, because the competition between FL formation and magnetic correlations should occur generally in frustrated electron systems. We expect that the reentrant behavior in the Mott transition found here will be observed experimentally in a variety of frustrated electron systems.

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[1] S. Kondo *et al.*, Phys. Rev. Lett. **78**, 3729 (1997).
[2] K. Takada *et al.*, Nature (London) **422**, 53 (2003).
[3] S. Lefebvre *et al.*, Phys. Rev. Lett. **85**, 5420 (2000).
[4] Y. Shimizu, K. Miyagawa, K. Kanoda, M. Maesato, and G. Saito, Phys. Rev. Lett. **91**, 107001 (2003).
[5] F. Kagawa, T. Itou, K. Miyagawa, and K. Kanoda, Phys. Rev. B **69**, 064511 (2004).
[6] T. Kashima and M. Imada, J. Phys. Soc. Jpn. **70**, 3052 (2001).
[7] H. Morita, S. Watanabe, and M. Imada, J. Phys. Soc. Jpn. **71** 2109, (2002).
[8] Y. Imai and N. Kawakami, Phys. Rev. B **65**, 233103 (2002).
[9] S. Onoda and M. Imada, Phys. Rev. B 67, 161102 (2003).
[10] S. Onoda and N. Nagaosa, J. Phys. Soc. Jpn. **72**, 2445 (2003).
[11] O. Parcollet G. Biroli, and G. Kotliar, Phys. Rev. Lett. **92**, 226402, (2004).
[12] S. Watanabe and M. Imada, J. Phys. Soc. Jpn. **73**, 3341 (2004).
[13] T. Watanabe, H. Yokoyama, Y. Tanaka, and J-i. Inoue, J. Phys. Soc. Jpn. **75**, 074707 (2006).
[14] H. Yokoyama, M. Ogata, and Y. Tanaka, J. Phys. Soc. Jpn. **75**, 114706 (2006).
[15] B. Kyung and A.-M. S. Tremblay, Phys. Rev. Lett. **97**, 046402 (2006).
[16] T. Koretsune, Y. Motome, and A. Furusaki, J. Phys. Soc. Jpn. **76**, 074719 (2007).
[17] B. Kyung, Phys. Rev. B **75**, 033102 (2007).
[18] A. Georges, G. Kotliar, W. Krauth and M. J. Rozenberg,

Rev. Mod. Phys. **68**, 13 (1996).
[19] A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **62**, R9283 (2000).
[20] G. Kotliar, S. Y. Savrasov, G. Pálsson, and G. Biroli, Phys. Rev. Lett. **87**, 186401 (2001).
[21] T. Ohashi, N. Kawakami, and H. Tsunetsugu, Phys. Rev. Lett. **97**, 066401 (2006).
[22] B. Kyung, G. Kotliar, and A.-M. S. Tremblay, Phys. Rev. B **73**, 205106 (2006).
[23] J. E. Hirsch and R. M. Fye, Phys. Rev. Lett. **56**, 2521 (1986).
[24] V. S. Oudovenko and G. Kotliar, Phys. Rev. B **65**, 075102 (2002).
[25] S. Moukouri and M. Jarrell, Phys. Rev. Lett. **87**, 167010, (2001).
[26] T. Maier, M. Jarrell, T. Pruschke, and M. H. Hettler, Rev. Mod. Phys. **77**, 1027 (2005).
[27] M. Jarrell and J. E. Gubernatis, Phys. Rep. **269**, 133 (1996).
[28] R. Zitzler N.-H. Tong, Th. Pruschke, and R. Bulla, Phys. Rev. Lett. **93**, 016406 (2004).